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DISTRIBUTION OF CESIUM-137 IN TREE CROP PRODUCTS COLLECTED FROM RESIDENCE ISLANDS IMPACTED BY THE U.S. NUCLEAR TEST PROGRAM IN THE MARSHALL ISLANDS

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DISTRIBUTION OF CESIUM-137 IN TREE CROP PRODUCTS COLLECTED FROM RESIDENCE ISLANDS IMPACTED BY THE U.S. NUCLEAR TEST PROGRAM IN THE NORTHERN MARSHALL ISLANDS

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The Marshall Islands Program at the Lawrence Livermore National Laboratory has completed a series of radiological surveys at Bikini, Rongelap, Utrōk, and Enewetak Atolls in the Marshall Islands designed to take a representative sample of food supplies with emphasis on determining ^{137}Cs activity concentrations in common food plants. Coconuts (*Cocos nucifera L.*) are the most common and abundant food plant, and provided a common sample type to characterize the level and variability of activity concentrations of ^{137}Cs in plant foods collected from different islands and atolls. Other dominant food types included *Pandanus* (*Pandanus* spp.) and breadfruit (*Actocarpus* spp.). In general, the activity concentration of ^{137}Cs in food plants was found to decrease significantly between the main residence islands on Bikini, Rongelap, Utrōk, and Enewetak Atolls. The mean activity concentration of ^{137}Cs measured in drinking coconut meat and juice was 0.72 (95% CI 0.68-0.77) and 0.34 (95% CI 0.30-0.38) Bq g^{-1}, respectively, on Bikini Island; 0.019 (95% CI 0.017-0.021) and 0.027 (95% CI 0.023-0.031) Bq g^{-1}, respectively, on Rongelap Island; 0.010 (95% CI 0.007-0.013) and 0.007 (95% CI 0.004-0.009) Bq g^{-1}, respectively, on Utrōk Island; and 0.002 (95% CI 0.0013-0.0024) and 0.002 (95% CI 0.001-0.0025) Bq g^{-1}, respectively, on Enewetak Island. High levels of variability are reported across all islands. These results will be used to improve the accuracy and reliability of predictive dose assessments, help characterize levels of uncertainty and variability in activity concentrations of fallout radionuclides in plant foods, and allow atoll communities to make informed decisions about resettlement and possible options for cleanup and rehabilitation of islands and atolls.

Keywords: Atmospheric Nuclear Weapons Testing · Bikini Atoll · Enewetak Atoll · Marshall Islands · ^{137}Cs activity concentration · coconut · *Pandanus* · breadfruit

Insert figure 1

Introduction

Between 1946 and 1958, the United States conducted 66 atmospheric nuclear tests in the Republic of Marshall Islands (Fig. 1). There were 42 nuclear tests conducted at Enewetak Atoll and 23 tests conducted at Bikini Atoll with one additional test conducted at high altitude (~26,200 m) about 97 kms west of Bikini [1]. The nuclear test program at Bikini and Enewetak led to significant local and regional fallout contamination of the northern Marshall Islands with a number of impacted communities still living in isolation from their ancestral homelands. The Lawrence Livermore National Laboratory (LLNL) under the auspices of the Office of Health and Safety, U.S. Department of Energy (DOE), continues to provide radiological monitoring of impacted atolls in the northern Marshall Islands in order to develop updated dose assessments for resettled and resettling populations, provide further understanding of the long-term behavior of fallout radionuclides in coral atoll ecosystems, and predict future change. There is also a continuing effort to evaluate the effectiveness of potential remedial options to help reduce or eliminate doses to island populations [2-3], and provide more direct support to those communities interested in exploring options for resettlement or who are simply interested in obtaining up-to-date information about gathering local foods from their home atolls to share with family or their community [4]. These studies follow a long history of U.S. agency as well as Marshall Islands Government sponsored studies on the health and ecological consequences of the nuclear testing program in the Marshall Islands. The ultimate aim of these studies is to provide guidance and assessments of radiological conditions on islands and atolls, and build a technical and scientific foundation for developing safe and sustainable resettlement programs [5-7].

Cesium-137 (^{137}Cs) is the highest contributor to nuclear test-related dose to people exposed to residual nuclear fallout contamination in the Marshall Islands. The radiological dose is dominated by the ingestion pathway. Recent studies show that the environmental loss-rate of ^{137}Cs from plants is more effective than radiological decay in accounting for future change in dietary intakes of ^{137}Cs [8]. These findings provide added assurances to resettled population groups that radiological conditions will likely remain at or below prescribed safety standards in radiological protection under existing land-use and exposure conditions, and may offer opportunity for other communities to advance plans for resettlement. The present study is partly motivated by the need to demonstrate that radiological conditions in the Marshall Islands are improving at an accelerated rate by the higher than expected loss-rate of ^{137}Cs from plants (and in the underlying soil) using repetitive sampling and analysis. Data are reported for ^{137}Cs activity concentrations in coconut, *Pandanus* fruit and breadfruit collected from the main residence island(s) of Bikini Atoll (Bikini and Enue Islands), Enewetak Atoll (Enewetak, Medrin and Japtan Islands), Rongelap Atoll (Rongelap Island) and Utrōk Atoll (Utrōk Island).

Experimental

Field Collection:

Since the early 2000s, field sample collections in the Marshall Islands have largely been formulated to mimic the use of island resources by collecting representative samples of local food supplies based on categories typically utilized by Marshall Islanders. Such collections are described under the LLNL pantry sampling program. Local Marshallese field crews were employed to help identify, collect and then process the field samples. The inflorescence of coconuts is normally studied from the ground. The preferred nut type is referred to as a 'drinking coconut'. Proper age determination for the coconuts can only occur after the husk is removed and the inner shell is examined but a 'good drinking coconut' typically contains a hard seed core, the juice chamber is full and turgid, and the meat has a white, soft texture. Younger or older nuts with more or less juice or copra nuts also form part of the local diet and were periodically collected. Coconut were usually collected from trees with the aid of an extension pole and immediately husked to ensure that nuts collected were at the optimum or preferred developmental stage. *Pandanus* fruit were the second most readily available food plant. If no ripe *Pandanus* fruit were available, then green fruit were often taken. After collection, the field samples were immediately placed in heavy duty plastic bags, the bags labeled, and the samples transferred to a central location for processing. Sample processing was done either aboard ship or within one of the field stations, and included the separation of coconut into coconut meat and juice. Care was taken to eliminate soil contamination and each sample was assigned a unique identifier (or ID Number). The ID number, date, island name, the number and condition of the fruit as well as other pertinent sample information were recorded in field log books.

Coconut, *Pandanus* fruit, breadfruit as well as any other plant foods were carefully washed as needed, patted dry with a tissue wipe, and the material diced into small sections. Processed samples were then placed in double, heavy duty (4 mil) plastic bags, weighed, sealed with vinyl tape, and labeled before being stored frozen for shipment to LLNL. Juice samples were typically strained and placed in 1 or 2-L plastic Nalgene bottles within a few hours of collection. Samples of coconut meat and juice usually consisted of a composite of 4 to 8 nuts of the same inflorescence. *Pandanus* fruit were split open and the individual *Pandanus* keys removed from the central fruit stalk. Only the more readily edible, bottom portion of individual *Pandanus* fruits were retained for sample analysis. Sample bags and juice bottles were labeled with the field ID number, date, site GPS location descriptor, and island name. No *Pandanus* or breadfruit samples were collected from Medrin or Japtan islands, and no breadfruit samples were collected from Utrōk Island.

Laboratory Analysis:

Sample fruits were dried to constant weight by lyophilization freeze-drying, homogenized by blending in a laboratory mixer, and then the material packed and sealed into a fixed geometry (typically metal tuna cans) for counting by high resolution gamma-spectrometry. Juice samples were allowed to thaw, acidified to pH 3-4 using hydrochloric acid, spiked with ^{134}Cs tracer, and cesium isotopes recovered by micro-extraction on 4 g of finely divided NH_4 molybdophosphate (AMP) powder. The AMP was recovered by centrifugation in 250 ml plastic Nalgene bottles. The bottles containing the AMP were then oven dried, placed on the face of a detector and counted by high resolution gamma-spectrometry. The amount of ^{137}Cs in the sample was qualified using an isotope dilution technique described elsewhere [9]. Blanks and quality control/quality assurance samples were prepared and gamma-counted in the same manner.

The Marshall Islands Program gamma-spectrometry facility houses up to 24 high-resolution gamma detectors coupled to a DEC VAXStation operating under Canberra Nuclear Data Systems software. Details concerning detector calibration and quality assurance procedures are also reported elsewhere [10-11]. All data are reported on a wet weight basis and normalized to a reference date of 1 January 2011 using an effective half-life of ^{137}Cs of 8.5 years [8].

Results and Discussion

The results of analyses of ^{137}Cs in food plants from the main residence islands on Bikini, Enewetak , Rongelap and Utrōk Atolls are summarized in Tables 1 and 2.

Insert Tables 1 and 2

The highest mean activity concentrations of ^{137}Cs in food plants were measured in copra meat samples with maximal levels of ^{137}Cs in food plants occurring on Bikini Island for all major food plant types with the exception of breadfruit. The highest level of ^{137}Cs in food plants where no remediation was used was observed on Bikini Island with maximal values typically exceeding the Codex Alimentarius guidelines (Codex 1994) for shipping of general food supplies across international borders. Food plants from the other islands all fall well below the Codex index. The difference in the mean activity concentration of ^{137}Cs measured in individual food plant types on Enewetak, Medren, and Japtan Islands on Enewetak Atoll was not statistically significant ($p > 0.05$) with the possible exception of ^{137}Cs in drinking coconut juice on Japtan and Medrin Islands ($p=0.03$). The activity concentrations of ^{137}Cs in most plant foods tended to show a highly skewed distribution mostly dominated by extreme values. The inter-island CV values (Coefficient-of-Variation) ranged between 51 and 157% for all food plants where 10 or more samples were analyzed. Studies from Bikini Island also show that there is a graded increase in the activity concentration of ^{137}Cs in both coconut meat and juice depending on the developmental stage (inflorescence) of the nuts [9]. In this way, the observed variation of activity concentration of ^{137}Cs in coconut juice from the same tree may vary by 3 to 5 fold depending on the type of nut selected. Disparate sampling of coconuts with different stages of development will therefore contribute to variability in the measurements. It should also be noted that coconut nuts collected from various islands varied in size with by far the smallest coconuts growing on trees from Rongelap Island. Bikini Island coconut trees tended to have the largest size nuts.

In general, the activity concentrations of ^{137}Cs in food plants decrease in significant incremental steps between Bikini, Rongelap, and Utrōk Islands, and Enewetak Atoll (including Enewetak, Medren and Japtan Islands). The typically ranking of ^{137}Cs activity concentration in plant foods on islands ordered highest to lowest was copra meat > *Pandanus*, breadfruit > copra juice > drinking coconut meat > drinking coconut juice. The mean activity concentration of ^{137}Cs measured in drinking coconut meat and juice was 0.72 (95% CI 0.68-0.77) and 0.34 (95% CI 0.30-0.38) Bq g^{-1}, respectively, on Bikini Island; 0.019 (95% CI 0.017-0.021) and 0.027 (95% CI 0.023-0.031) Bq g^{-1}, respectively, on Rongelap Island; 0.010 (95% CI 0.007-0.013) and 0.007 (95% CI 0.004-0.009) Bq g^{-1}, respectively, on Utrōk Island; and 0.002 (95% CI 0.0013-0.0024) and 0.002 (95% CI 0.001-0.0025) Bq g^{-1}, respectively, on Enewetak Atoll (see Figure 2). The intra-island variability in measured ^{137}Cs activity concentration across the other food plants (with exception of breadfruit on Bikini Island) follows a similar trend consistent with previous findings on inter-plant concentration ratios (ICPRs) [12].

Insert figure 2

The total number of *Pandanus* fruit and breadfruit collected during this study tended to be lower than for coconut products but still provided useful information. As with drinking coconut and copra products, the activity concentration of ^{137}Cs in *Pandanus* fruit was highest on Bikini Island (mean = 0.99 Bq g^{-1}, full range = 0.27-2.3 Bq g^{-1}). In breadfruit, however, the mean ^{137}Cs activity concentration observed on Rongelap Island (mean = 0.20 Bq g^{-1}, N=8, full range = 0.089-0.466 Bq g^{-1}) was twice the value observed at Bikini Island (mean = 0.12 Bq g^{-1}, N=9, full range = 0.046-0.31 Bq g^{-1}). In this case, the breadfruit on Bikini is being consumed by local workers living on the island, and the breadfruit trees were treated with potassium fertilizer to help reduce or eliminate the uptake of ^{137}Cs from the food. Based on large-scale field experiments on Bikini, the addition of 2000 kg potassium per hectare is expected to reduce ^{137}Cs concentrations in coconut meat on Bikini by a factor of about 20 [2]. In this case, effectiveness of potassium treatment of breadfruit on Bikini relative to measurements of ^{137}Cs in untreated trees on Rongelap appear to scale proportionally with expectations. Moreover, the activity concentrations of ^{137}Cs on food plants from Enue Island on Bikini Atoll were also consistently lower than those observed on Bikini Island. Average concentrations of ^{137}Cs in surface soils on the interior of Enue Island are about a factor of 10 times less than those observed on Bikini. Nonetheless, the observed difference in activity concentration of ^{137}Cs in coconut products on Enue and Bikini Islands can be partly attributed to historical efforts to fertilize Enue Island with potassium.

CONCLUSIONS

Approximately 85-90% of the radiation dose delivered to humans exposed to residual nuclear fallout contamination in the Marshall Islands is derived from ingestion of ^{137}Cs contained in locally grown food plants [1]. Radiological doses from the ingestion pathway will scale directly with the total intake of radionuclides. Previous assessments show that dietary intakes of ^{137}Cs are dominated by consumption of locally grown tree-crop foods, primarily from consumption of coconut, *Pandanus* fruit and breadfruit. An extensive effort has therefore been made to develop databases on ^{137}Cs activity concentrations in these types of food plants in order to help refine existing dose assessments, predict future change in radiological conditions, and assess any future needs for cleanup and rehabilitation of islands or atolls. The present study supports the development of updated databases for use in radiological assessments which include uncertainty and variability in estimates of potential ingestion doses from ^{137}Cs.

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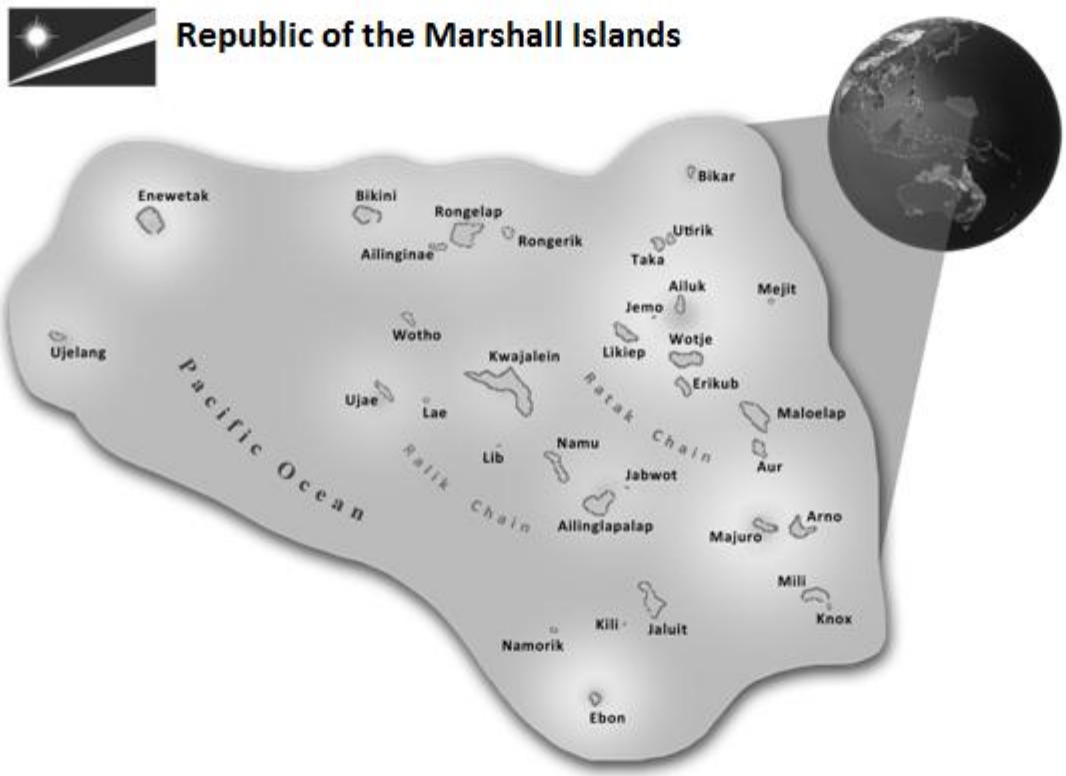

Figure 1. Map showing location of Bikini, Enewetak, Rongelap and Utrōk Atolls in the northern Marshall Islands

Table 1. ^{137}Cs activity concentration in food plant products from residence islands of Bikini and Rongelap Atolls

Island	Food Plant	^{137}Cs (Bq g^{-1}, wet wt.) N	Mean	Median	Range
Bikini Atoll					
Bikini	Coconut Meat	279	0.72 ± 0.41	0.653	0.043 - 2.7
	Coconut Juice	184	0.34 ± 0.28	0.250	0.022 - 1.5
	Copra Meat	158	1.11 ± 0.77	0.940	0.049 - 3.6
	Copra Juice	94	0.76 ± 0.59	0.611	0.042 - 2.4
	Pandanus	18	0.99 ± 0.65	0.788	0.27 - 2.3
	Breadfruit	9	0.120 ± 0.09	0.267	0.046 - 0.31
Enue	Coconut Meat	12	0.031 ± 0.038	0.023	0.004 - 0.147
	Coconut Juice	12	0.017 ± 0.022	0.005	0.002 - 0.081
	Copra Meat	9	0.09 ± 0.10	0.049	0.012 - 0.359
	Copra Juice	7	0.042 ± 0.029	0.030	0.009 - 0.102
	Pandanus	2	0.030 ± 0.007	0.030	0.022 - 0.037
	Breadfruit	2	0.078 ± 0.004	0.078	0.075 - 0.082
Rongelap Atoll					
Rongelap	Coconut Meat	245	0.019 ± 0.020	0.013	0.002 - 0.143
	Coconut Juice	306	0.027 ± 0.033	0.018	0.000 - 0.364
	Copra Meat	62	0.061 ± 0.062	0.043	0.006 - 0.396
	Copra Juice	62	0.047 ± 0.038	0.036	0.006 - 0.204
	Pandanus	26	0.090 ± 0.081	0.053	0.009 - 0.339
	Breadfruit	8	0.20 ± 0.13	0.149	0.089 - 0.466

Table 2. ^{137}Cs activity concentration in food plant products from residence islands of Enewetak and Utrōk Atolls

Island	Food Plant	^{137}Cs (Bq g^{-1}, wet wt.)			
		N	Mean	Median	Range
Enewetak Atoll					
Enewetak	Coconut Meat	35	0.002 ± 0.003	0.002	0.0003 - 0.013
	Coconut Juice	55	0.002 ± 0.003	0.001	0.0000 - 0.018
	Copra Meat	39	0.006 ± 0.008	0.002	0.0003 - 0.045
	Copra Juice	37	0.004 ± 0.004	0.002	0.0001 - 0.019
	Pandanus	6	0.010 ± 0.005	0.004	0.005 - 0.020
	Breadfruit	15	0.009 ± 0.013	0.007	0.002 - 0.051
Medrin	Coconut Meat	9	0.001 ± 0.001	0.001	0.0000 - 0.003
	Coconut Juice	10	0.001 ± 0.001	0.001	0.0002 - 0.004
	Copra Meat	16	0.007 ± 0.006	0.005	0.0008 - 0.027
	Copra Juice	17	0.005 ± 0.004	0.005	0.0007 - 0.015
Japtan	Coconut Meat	13	0.003 ± 0.002	0.003	0.0003 - 0.009
	Coconut Juice	12	0.003 ± 0.002	0.003	0.0003 - 0.008
	Copra Meat	13	0.006 ± 0.006	0.004	0.0000 - 0.022
	Copra Juice	13	0.005 ± 0.003	0.005	0.0003 - 0.011
Utrōk Atoll					
Utrōk	Coconut Meat	12	0.010 ± 0.005	0.006	0.002 - 0.019
	Coconut Juice	13	0.007 ± 0.004	0.010	0.002 - 0.015
	Copra Meat	6	0.016 ± 0.008	0.015	0.004 - 0.032
	Copra Juice	6	0.01 ± 0.007	0.008	0.003 - 0.024
	Pandanus	2	0.011 ± 0.001	0.011	0.010 - 0.012

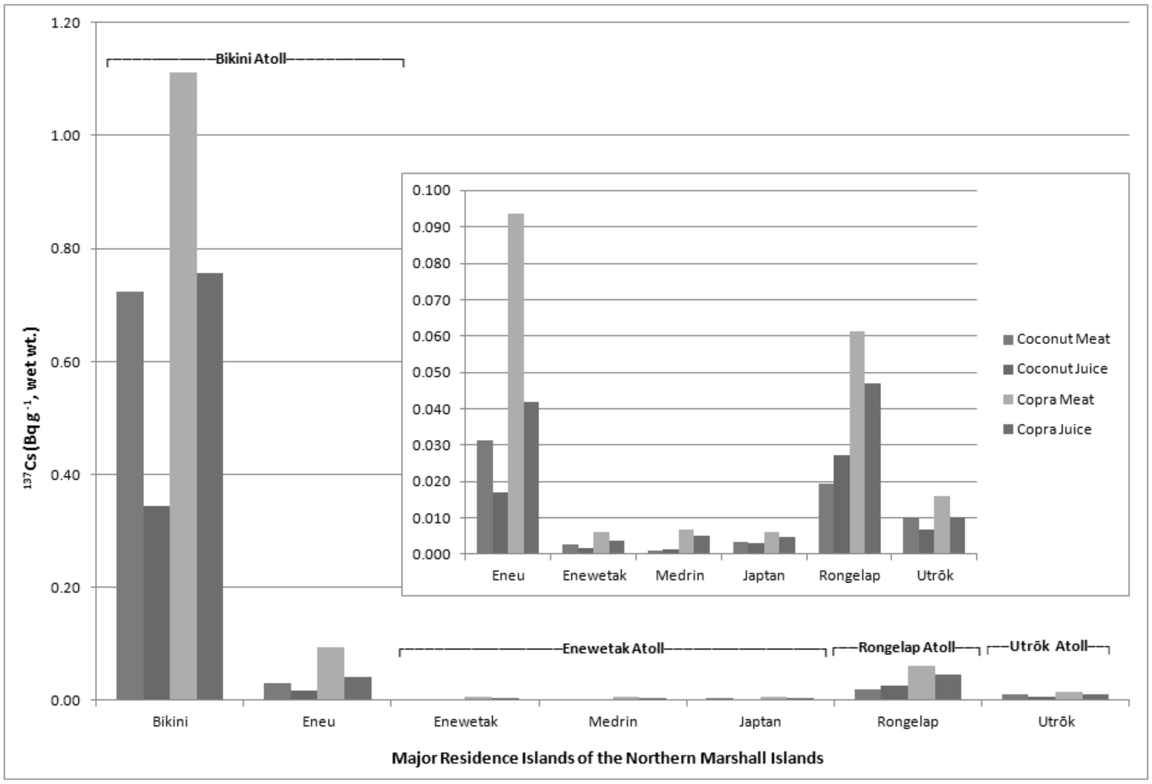

Figure 2: Mean ^{137}Cs activity concentration in coconut products (coconut meat and juice, copra meat and juice) sampled from the major residence islands in the Northern Marshall Islands including Bikini Island and Eneu Island (Bikini Atoll); Enewetak, Medrin, and Japtan Islands (Enewetak Atoll); Rongelap Island (Rongelap Atoll), and Utrōk Island (Utrōk Atoll).